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(54) RADIATION SHIELDING

(71) We, CHEMTREE CORPORATION, a corporation organized under the laws of the State of Delaware, having a principle place of business at Central Valley, State of New York, United States of America, (Assignee of WILLIAM CORNELIUS HALL and JOHN MERRIMAN PETERSON), do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to improvements in radiation shielding and more particularly to shielding with high efficiency in removing neutrons, and also to shielding providing for degradation of neutrons and attenuation of gamma radiation resulting from neutron capture.

Interest in radiation shielding material is increasing at an accelerated rate in a wide variety of uses, ranging from atomic reactors to space exploration. Rapid advances have been made in both theory and technology. The large number of nuclear power plants already in use and under construction, together with those projects on the drawing board, or announced as part of future planning, clearly demonstrates that expansion in this area is growing rapidly. The expanding usage of radioisotopes is well known, and X-ray machines have acquired a large multiple usage in medicine where high energy therapy is employed against cancer. X-ray machines also have expanded into industry and they are a common tool for non-destructive testing and inspection work. In addition to the 300 billion electron volt accelerator which the United States Atomic Energy Commission plans to construct, several industrial companies are manufacturing smaller size particle accelerators, used extensively in research projects and industrial processes. The instrumentation field also sometimes requires special types of shielding materials. Each application presents different radiation problems, and consequently the types of radiation shielding material for different applications vary widely in basic specifications.

Much attention has been given to atomic reaction shielding where bulk shielding is practical. This application usually emphasizes shielding material with low cost ingredients and good structural properties. Generally, these factors are achieved at the expense of mass and volume considerations.

However, in the field of certain specialized research, especially in the area of space exploration, cost and structural properties are much less important than low weight and small volume. For these situations, attenuation efficiencies for the particular radiations involved are of prime importance. The solution of such radiation problems is one feature of this invention.

Selection of shielding material for various types of radiation must consider the modes of decay for the initial reaction. These modes may involve one or more of the following: alpha particles, negative beta particles, positive beta particle, K-electron capture, L-electron capture, isomeric transition, neutron capture (or neutron emission), proton capture (or proton emission), and spontaneous fission. Subsequent radiations, resulting from interaction of primary radiation with other matter which it strikes, may include one or more of the following: gamma ray, prominent internal conversion electron, "delayed" neutron, "delayed" alpha particles, "delayed" beta particle.

It has been found that metals with atomic numbers 57 through 71, inclusive, and compounds of such metals have special shielding advantages for certain situations demanding high attenuation efficiencies. Specifically, attenuation efficiencies of a high order are achieved for a variety of mixed flux situations by the utilization of homogeneous materials containing dysprosium, gadolinium, europium and samarium with atomic numbers 66, 64, 63 and 62, respectively.

Comparison of two of these materials, dysprosium and gadolinium, with commonly

used shielding materials such as boron and carbon with atomic numbers of 5 and 6 respectively, illustrates the following improvements in certain attenuation efficiencies: The scatter cross-section to degrade neutron energies for dysprosium is 100 barns, compared with 4.8 barns for carbon; the thermal neutron capture cross-section to remove neutrons for dysprosium is 0.95×10^3 barns compared with 0.76×10^3 for boron; and the thermal neutron capture cross-section to remove neutrons for gadolinium is 4.6×10^4 barns compared with 7.6×10^2 for boron. The mass of both dysprosium and gadolinium is sufficient for the attenuation of considerable photon energy resulting from inelastic scatter and from some of the captures.

Under certain circumstances improved attenuations may be had by utilizing compounds of dysprosium and gadolinium rather than the pure metals. For instance dysprosium hydride provides very superior elastic scatter of neutrons by hydrogen where the enutron energy is largely lost by collision, as compared to inelastic scatter where the neutrons are replaced by photons which then need to be attenuated. For the same reason it may be desirable to utilize gadolinium hydride. Gadolinium tungstate has large usefulness where neutrons have been degraded to thermal energy and there is considerable photon energy in the flux. In such cases, gadolinium captures thermal neutrons, gadolinium and the tungsten both attenuate photons. Tungsten has very high linear efficiency because of its very high density.

While these metals with atomic numbers 57 through 71, inclusive, alone or in combination, may be held in position in containers, they may be dispersed in polyolefins (homo- or co-polymers) to produce a product with better structural characteristics. These polyolefin compositions may be processed on various types of equipment such as injection, compression, transfer and blow molding machinery, as well as by calendering or extrusion techniques.

The use of a polyolefin base, such as polyethylene, has the particular advantages of ease with which ingredients used for formulation may be mixed in widely different proportions, and of desirable characteristics in the physical and shielding properties found in the finished products.

Properties, useful in many applications, furnished by this base polyolefin material include its light weight, good flexibility, high degree of toughness and chemical resistance. The hydrocarbon itself complements the shielding effectiveness of the included ingredients as will be shown later in specific formulations.

"Linear" or "high density" polyethylene has several special characteristics not possessed by conventional polyethylene including a wider temperature range of stability, shorter mold cycle time, and greater stiffness, which adds to structural strength. In neutron shielding, it has a greater number of hydrogen atoms per unit volume and this contributes to the composite neutron capture efficiency. Heat treatment at higher temperatures of the molded material further improves thermal stability and structure. Consequently, linear polyethylene has advantages when used as a base material.

The metals and compounds of elements with atomic numbers 57 through 71, inclusive, may be dispersed in the polyolefin base in standard equipment of the Banbury mixer or roll-mill type. Preferably the metallic elements are dispersed in the polyolefin to an extent of up to 10% by volume thereof.

Examples of improved shielding products containing elements with atomic numbers 57 through 71, inclusive, are described in the following formulations:

Example 1

Two volumes of dysprosium powder were dispersed uniformly into one volume of high density polyethylene (previously softened thermally to a dough consistency), on a roll-mill, and thence injection molded into a test specimen and subsequently cooled. The final product had good structural strength for shielding purposes and shows a high degree of attenuation improvement of this product over polyethylene itself and over other common shielding materials with low atomic numbers.

Example 2

One volume dysprosium powder and one volume gadolinium powder were dispersed uniformly into one volume of polyethylene (pellets) heated to a dough consistency, on a roll-mill. The softened mass was pressure molded into a test specimen and showed satisfactory structural strength upon cooling as well as excellent attenuation efficiencies.

Example 3

One volume dysprosium oxide powder and one volume polyethylene (powder) were thoroughly mixed and the mixture further blended on a roll-mill at approximately 280°C for seven minutes. The product was molded into a specimen of desired shape and cooled. The specimen has good structural characteristics and superior attenuation properties.

Example 4

One volume of gadolinium hydride and one volume linear polyethylene were intimately mixed on a roll-mill at a temperature sufficiently high to give a workable consistency. The composite was molded to desired shape and cooled.

The gadolinium hydride content may be increased to the fullest extent possible with

both increased performance and cost, and contrarily it may be decreased with attendant loss of efficiency and cost.

EXAMPLE 4A

Dysprosium hydride in equal volume was substituted for gadolinium hydride in Example 4.

As dysprosium is about six times as efficient as carbon on a mass basis and this compound has about the same hydrogen content by weight of polyethylene, there is a considerable net gain in neutron moderation over solid polyethylene, roughly estimated to be about 25% on a mass basis and 125% on a linear basis. It is estimated that a 0.45 thickness of polyethylene with dysprosium hydride equals 1.00 thickness of polyethylene for neutron degradation and that 0.45 thickness of polyethylene with dysprosium hydride has a mass of 0.8 as compared to 1.0 for polyethylene.

It is calculated that the thermal neutron capture is improved by a power of 10^3 and that the gross neutron capture is improved by about 10^2, the hydrogen content remaining about the same.

It thus appears that polyethylene with dysprosium hydride dispersed through it is an improved neutron shield.

EXAMPLE 5

Three volumes of dysprosium powder, one volume of gadolinium powder and two volumes of linear polyethylene were blended on a roll-mill. The resultant product was molded and subsequently cooled. The resultant sample provides in a single shielding material elements effective for degrading or moderating high energy neutrons by both elastic and inelastic scatter, for thermal neutron capture, and for the attenuation of significant photon energy resulting from inelastic scatter of neutrons and from the capture of some of the neutrons. This composition is especialy effective for use in nuclear radiation shielding situations involving a high initial flux of fission spectrum neutrons or neutrons with even higher energies. Its effectiveness is improved further by lamination with high Z material behind it for photon attenuation.

EXAMPLE 6

One volume of gadolinium tungstate was blended thoroughly with one volume of polyethylene (pellets) on a roll-mill, molded and then cooled. The presence of tungsten in the formulations improves the shielding efficiencies for certain uses, as previously described.

WHAT WE CLAIM IS:—

1. A radiation shielding composition comprising one or more metallic elements with an atomic number from 57 to 71 inclusive or compounds of said elements homogeneously disposed in a polyolefin binder.

2. A composition as claimed in claim 1 in which the dispersed metal is dysprosium, gadolinium and/or europium, in powder form.

3. A composition as claimed in claim 1 in which the dispersed material is a compound of dysprosium, gadolinium and/or europium.

4. A composition as claimed in claim 1 in which the dispersed metals are in the form of their hydrides.

5. A composition as claimed in claim 1 in which the dispersed metal is in the form of gadolinium tungstate.

6. A composition as claimed in any of claims 1 to 5 comprising a polyolefin of doughy consistency with the dispersed materials homogeneously distributed therein and set to form a structure of predetermined form.

7. A composition as claimed in any of claims 1 to 6 in which the metallic elements dispersed therein make up no more than 10% by volume thereof.

8. A composition as claimed in any of claims 1 to 7 in which the polyolefin is polyethylene.

9. A composition as claimed in claim 8 in which the plastic material is linear (high density) polyethylene.

10. A composition according to claim 1 substantially as hereinbefore described with particular reference to any of the foregoing Examples.

11. A radiation shield comprising a composition as claimed in any of claims 1 to 10 when molded to form a solid structure.

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